DISPLACEMENT CHROMATOGRAPHY METHODS FOR PRODUCING BATTERY-GRADE LITHIUM, COBALT, NICKEL, AND MANGANESE FROM BLACK MASS AND MINERAL CONCENTRATES

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Abstract

Lithium-ion batteries are cornerstones of electric vehicles, grid storage, and consumer electronics. They rely on lithium (Li), cobalt (Co), nickel (Ni), and manganese (Mn) as their essential active materials. Yet the U.S. imports over 97% of these battery-grade compounds, reflecting limited domestic refining capacity and exposing the nation to significant supply chain risks. These vulnerabilities are compounded by geopolitical, regulatory, and resource availability constraints that threaten supply disruptions. Any shortfall in their supply may fail to meet their growing demand. Therefore, novel methodologies for sourcing these critical materials, along with identifying alternative substitutes and implementing advanced recycling and reuse processes to maximize their utility, are needed.

In this dissertation, the successful purification of Li, Co, Ni, and Mn from diverse complex feedstocks, including mineral ores and black masses derived from the grinding and sieving of electric vehicle batteries, is presented. The purification is chromatography-based and was initially designed for batch chromatography but later extended to continuous chromatography. The approach for batch chromatography integrates an innovative constant-pattern design method tailored for non-ideal displacement systems, enabling the estimation of optimal operating parameters without trial and error. These parameters ensure the separation of individual pure components with predefined target purities and yields while maximizing sorbent utilization. Simulations and experiments were conducted to validate the approach, resulting in the production of individual high-purity (>99.5%) Li, Co, Ni, and Mn products at yields exceeding 99% from cathode materials, black masses, and mineral concentrates. All the experiments are conducted at room temperature, with the pressure drop maintained below 100 psi, leading to low energy consumption. Additionally, the influence of the displacer concentration, column dimensions, zone

configurations, and sorbent particle size on productivity was investigated and optimized to maximize the kilograms of pure (>99.5%) target products per cubic meter of sorbent per day.

Building on these results, simulated moving bed (SMB) chromatography systems were developed, in which the separation zones are coupled with stripping and re-equilibration zones to enable continuous separation of Li, Ni, Co, and Mn salts. These continuous operations are designed using standing-wave theory and further refined through the introduction of preloading steps, which allow the process to reach cyclic steady state within a single cycle. In these configurations, the fast-moving solute is effectively recovered in the Raffinate, whereas the more strongly retained solutes are temporarily held within the system and subsequently withdrawn in the Extract or associated stripping zone.

Collectively, the methods developed in this work offer a pathway to move Li, Co, Ni, and Mn supply chains away from a linear "mine-use-dispose" model toward a more sustainable circular economy, thereby supporting the long-term viability of critical technologies.